412 Rec'd PCT/PTO 31 AUG 2000

FORM PTO-1390 (REV 12-29-99) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER: WEB 0039 PA

U.S. APPLICATION NO. (If known, see 37 CFR 1.5): 09/623588

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/EP99/01361	March 3, 1999	March 4, 1998

TITLE OF INVENTION: METHOD OF CARRYING OUT CHEMICAL REACTIONS IN A MICROREACTOR AND SUCH A MICROREACTOR

APPLICANT(S) FOR DO/EO/US: Ehrfeld et al

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
   a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☒ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). UNSIGNED
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
    ☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information: Certificate of Express Mail filing

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U.S. APPLICATION NO. (if known, see 37 CFR 1.5)	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
09/623588	PCT/EP99/01361	WEB 0039 PA

17. [XX] The following fees are submitted:

	CALCULATIONS	PTO USE ONLY
BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :		
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO **$970.00**		
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO **$840.00**		
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO **$690.00**		
International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) **$670.00**		
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) **$96.00**		
ENTER APPROPRIATE BASIC FEE AMOUNT =	$ 840.00	
Surcharge of **$130.00** for furnishing the oath or declaration later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(e)).	$ 00	

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	19 - 20 =	0	X **$18.00**	$ 00	
Independent claims	2 - 3 =	0	X **$78.00**	$ 00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)		0	+ **$260.00**	$ 00	

TOTAL OF ABOVE CALCULATIONS =	$ 840.00	
Reduction of 1/2 for filing by small entity, if applicable. A Small Entity Statement must also by filed (Note 37 CFR 1.9, 1.27, 1.28).	$ 00	
SUBTOTAL =	$ 840.00	
Processing fee of **$130.00** for furnishing the English translation later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(f)). +	$ 00	
TOTAL NATIONAL FEE =	$ 840.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). **$40.00** per property +	$ 00	
TOTAL FEES ENCLOSED =	$ 840.00	
	Amount to be refunded:	$
	charged:	$

a. [XX] A check in the amount of $ 840.00 to cover the above fees is enclosed.

b. [ ] Please charge my Deposit Account No. ________ in the amount of $________ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. [ ] The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. ________. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Richard A. Killworth
Killworth Gottman Hagan & Schaeff, LLP
One South Main Street, Suite 500
One Dayton Centre
Dayton, OH 45402-2023

Richard A. Killworth

SIGNATURE:

Richard A. Killworth

NAME

26,397

REGISTRATION NUMBER

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Application of

Applicant : Ehrfeld et al
Title METHOD OF CARRYING OUT CHEMICAL REACTIONS IN A MICROREACTOR AND SUCH A MICROREACTOR
Docket : WEB 0039 PA

BOX PCT
Assistant Commissioner
for Patents
Washington, DC 20231

Sir:

PRELIMINARY AMENDMENT

Prior to examination of the above-identified patent application, please amend the application as follows:

IN THE SPECIFICATION

In all instances, please change the spelling of "minimise" to - -minimize- -.

In all instances, please change the spelling of "minimisation" to - -minimization- -.

Page 1, line 6, delete "Description" and insert therefor - - Background and Summary of the Invention - -.

Page 13, between lines 6 and 7, insert - -Brief Description of the Drawings- -.

Page 14, between lines 2 and 3, insert - -Detailed Description- -.

IN THE CLAIMS

Please cancel claims 1-19 and insert therefor the following new claims 20-38:

20. A method of carrying out chemical reactions in a microreactor, in which one or more educt streams are guided to at least one reaction region and in which one or more product streams are guided from the reaction region in counterflow relationship with the educt stream or streams and in thermal contact therewith, characterized in that to minimize heat losses from the reaction region to the surrounding area the educt stream or streams and the product stream or streams are guided spirally or radially in at least one plane respectively to or from the reaction region arranged in a central region of the microreactor.

21. A method as set forth in claim 20 characterized in that a plurality of educt and product streams are respectively guided radially to and from the central region in alternate sectors of a space which is of an annular configuration in cross-section and which surrounds a cylindrical central region, wherein the central region and/or regions of the sectors serves or serve as the reaction region.

22. A method as set forth in claim 20 characterized in that a plurality of educt and product streams are guided radially respectively to and from a plurality of reaction regions which are fluidically separated from each other and which are disposed in mutually adjacent relationship and in a central region of the microreactor, wherein in each case at least one educt and product stream are guided in a counterflow reactor unit which has at least one respective reaction region.

23. A method as set forth in claim 22 characterized in that a heat exchange medium is passed around the counterflow reactor units.

24. A method as set forth in claim 21 characterized in that the educt streams and/or the product streams are fed and discharged respectively by way of an outer space of an annular configuration.

25. A method as set forth in claim 20 characterized in that the educt stream or streams and/or the product stream or streams are guided in passages of a width in at least one region of < 2 mm.

26. A microreactor for carrying out chemical reactions comprising one or more feed means for educts and one or more discharge means for products and at least one reaction region connected to the feed and discharge means, wherein the discharge means are arranged in counterflow relationship with the one or more feed means and are in thermal contact therewith, characterized in that to minimize heat losses from the reaction region to the surrounding area the feed means and the discharge means or the plurality of feed means and the plurality of discharge means are arranged spirally or radially in at least one plane around the reaction region which is in a central region of the microreactor.

27. A microreactor as set forth in claim 26 characterized by a cylindrical central region and a space which is of an annular configuration in cross-section and which surrounds it and which is divided into sectors,

wherein alternate sectors of the space of an annular configuration serve as feed and discharge means respectively,

wherein the sectors serving as a feed means are connected by way of the central region to the respective sectors serving as a discharge means, and

wherein the central region and/or regions of the sectors serves or serve as a reaction region .

28. A microreactor as set forth in claim 26 characterized in that a plurality of feed and discharge means are arranged radially in at least one plane around a plurality of reaction regions which are fluidically separated from each other and which are disposed in mutually adjacent relationship in a central region of the microreactor, wherein at least one respective feed means and discharge means and at least one reaction region are contained in a counterflow reactor unit.

29. A microreactor as set forth in claim 28 characterized in that the counterflow reactor units each have a counterflow tube including an outer tube closed at one end and an inner tube spaced coaxially in relation to the outer tube, wherein the inner tube serves as a feed or discharge means and the region between the outer and the inner tube serves as a discharge or feed means respectively, and wherein the transitional region between the feed and the discharge means in the region of the closed end of the outer tube serves as a reaction region.

30. A micro reactor as set forth in claim 28 characterized in that the counterglow reactor units are arranged in two or more planes radially around a cylindrical central region.

31. A micro reactor as set forth in claim 28 characterized in that the counterglow reactor units are arranged in the space radially around the central spherical region.

32. A micro reactor as set forth in claim 28 characterized in that the counterglow reactor units each have two mutually spaced plate-shaped elements forming a passage serving as a feed or discharge means, and a plate-shaped element which surrounds the two plate-shaped elements in a U-shape and which forms therewith a respective passage serving as a discharge or feed means, wherein the transitional region between the feed and the discharge means in the region of the bend of the U-shaped element serves as a reaction region.

33. A micro reactor as set forth in claim 28 characterized in that the central region having the counterglow reactor units is adapted for a heat exchange medium to pass there through.

34. A micro reactor as set forth in claim 27 characterized in that the sectors or the counterglow reactor units are surrounded by an outer space which is of an annular configuration in cross-section and which serves as a discharge collector or feed distributor and which is connected in the radial direction to the discharge means and the feed means respectively.

35. A micro reactor as set forth in claim 34 characterized in that the outer space which is of an annular configuration and which serves as a discharge collector 59a) or feed distributor is surrounded by a second outer space which is of an annular configuration in cross-section and which serves as a feed distributor or discharge collector respectively and which is connected in the radial direction to the feed means (43; 56) and the discharge means respectively.

36. A micro reactor as set forth in claim 27 characterized by a main discharge means or main feed means which is arranged above and/or below the space which is of an annular configuration and which is divided into sectors and/or the first and/or second outer space which is of an annular configuration and which for that purpose is connected in the axial direction to the sectors serving as the discharge means or feed means respectively or the discharge collector or the feed distributor respectively.

37. A micro reactor as set forth in claim 26 characterized in that the feed and/or discharge means are in the form of passages in at least one region of a width of <2 mm.

38. A micro reactor as set forth in claim 26 characterized by catalytically active material which is possibly applied to a carrier, in the reaction region.

IN THE ABSTRACT

Please insert the following abstract:

- -Abstract of the Disclosure

Microreactors are distinguished in particular by a high selectivity and yield of the chemical reactions carried out therein. The high surface-to-volume ratio however results in high heat losses from the reaction region to the surrounding area even in counter-flow guidance of the educt and product steams whereby those microreactors are limitedly suitable for use for chemical reactions in high temperatures. The object of minimizing such heat losses is attained with a method in which the educt and product streams are guided spirally or radially respectively to and from the reaction region disposed in a central region of the microreaction The reaction region is therefore surrounded in at least one plane by the educt and product streams which are guided in counterglow relationship with each other, so that the reaction heat generated is for a large part fed to the reaction region again. As a result the area of use of microreactors is decisively expended with regard to reactions at high temperature. A corresponding reactor in different configurations is also described.- -

This amendment is being made to place the application in better condition for examination and to delete the multiple claim dependency.

Respectfully submitted,

KILLWORTH GOTTMAN HAGAN &
SCHAEFF, L.L.P.

By Richard A. Killworth
Richard A. Killworth
Reg. No. 26,397

One Dayton Centre
One South Main Street, Suite 500
Dayton, Ohio 45402-2023

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Method of carrying out chemical reactions in a microreactor and such a microreactor

Description

The invention concerns a method of carrying out chemical reactions in a microreactor, in which one or more educt streams are guided to at least one reaction region and in which one or more product streams are guided from the reaction region in counterflow relationship with the educt stream or streams and in thermal contact therewith. The invention further concerns a microreactor for carrying out chemical reactions having one or more feed means for educts and one or more discharge means for products and at least one reaction region connected to the feed means and discharge means respectively, wherein the discharge means are arranged in counterflow relationship with the one or more feed means and in thermal contact therewith.

By virtue of the high surface-to-volume ratio, microreactors permit fast precise adjustment of reaction conditions and thus optimisation of yield and selectivity. As only small amounts are reacted for each reaction region, reactions involving explosive and/or toxic substances can also be carried out, without extensive safety precautions. That permits for example on-site production of small amounts, in particular of substances whose production and/or transportation on a large scale are not practicable.

DE 39 26 466 A1 discloses a microreactor for carrying out chemical reactions with considerable heat production, in which substance, reaction and heat conduction takes place in plate-like elements which are arranged in mutually superposed layered relationship and which are connected and run through by a system comprising grooves produced by machining. The microreactor itself can be made from a catalyst material.

WO 95/30476 sets forth a method of carrying out chemical reactions. Because the educts are divided into fluid threads, the educts pass in closely adjacent relationship in the form of free jets into a space which serves as a mixing and reaction chamber where they are mixed and react due to diffusion and turbulence. The advantage in that respect is that the division of the educts into fluid threads means that they are rapidly homogeneously mixed whereby a reaction takes place with few consequential and by-products. Implementation of heterogeneously catalysed gaseous phase reactions is not readily possible using that method.

A crucial disadvantage of known microreactors is that a high level of heat loss occurs by virtue of the high surface-to-volume ratio which permits specific, targeted and rapid adjustment of the reaction conditions. Particularly in the case of reactions which take place at elevated temperatures, that requires a continuous supply of relatively large amounts of heat in order to compensate for the heat losses by way of the microreactor housing and by way of the product stream which leaves the reactor.

On a large scale this problem scarcely arises by virtue of the fact that the actual reaction region is thermally easy to insulate and the reaction heat from the product stream is easily recovered by way of heat exchangers. Multi-stage design configurations of that kind however cannot be readily transferred to microreactors and in addition would nullify the advantage of compact arrangements.

A large-scale reactor of that kind for carrying out exothermic catalytic oxidation reactions under conditions which are as isothermal as possible is described in German laid-open application (DE-OS) No 2 016 614. In the cylindrical reactor, the gas mixture to be reacted is fed axially by way of a central gas feed means and is passed radially into circular-ring or annular sectors filled with catalyst material. The reaction takes place in the annular sectors, in which respect alternate annular sectors are in the form of plate heat exchangers through which a cooling fluid flows in the

axial direction. The reaction heat absorbed by the cooling fluid can be given off to the gas mixture to be reacted, for example by way of a further heat exchanger, in order to achieve a required initial temperature. The reaction products which flow radially out of the annular sectors are brought together by way of an annular space which is in the form of a collector or manifold and which surrounds the annular sectors. This configuration which includes at least one heat exchanger would however result in high levels of heat loss in the event of its being transferred to microreactor scales, by virtue of the complex structure involved.

US No 5 405 586 also sets forth a cylindrical reactor having annular sectors as reaction regions and heat exchangers. Each second annular sector has a catalytically active material through which the flow is radially from the outside inwardly, wherein the educts are supplied axially from above and the products are discharged axially downwardly. Alternate sectors have a heat exchanger medium flowing therethrough radially from the inside outwardly so that the temperature of the catalyst-filled reaction regions can be precisely adjusted.

A further large-scale reactor for carrying out catalytic reactions, in which the reaction regions are in close contact with a heat exchanger, is described in German patent specification No 942 805. Disposed between the turns of a spiral heat exchanger is catalytically active material through which a flow passes in transverse relationship with the heat exchanger medium which flows in a spiral configuration.

A similar reactor is described in the Abstract for JP 3-26326. The gases to be reacted are passed from the inside outwardly in a spirally wound catalytic layer while in the intermediate spaces which are also spirally wound, a heating medium is firstly supplied axially and then passed outwardly in a spiral configuration.

The Abstract for JP 61-118132 describes a cylindrical reactor in which the flow is through a catalytic material disposed between spirally wound plates, in axial relationship with respect to the axis of the spiral. A heat exchanger medium flows transversely with respect to the educt flow

in the intermediate spaces, which also extend in a spiral configuration, of the spirally wound plates. In this case also the reaction regions are in direct contact with a heat exchanger.

German patent specification No 214 788 sets forth a closed spiral counterflow cooler for heat exchange between two fluids which are separated from each other. No provision is made for use as a reactor.

Large-scale reactors of this kind are optimised in regard to the option of specifically and targetedly adjustable isothermal reaction implementation. Minimisation of heat losses of the reaction regions directly to the surrounding area or indirectly by way of the discharged products is however not an aim here. By virtue of the heat losses which are involved with the altered surface-to-volume ratios, the application of such reactor design configurations to the scales of microreactors is scarcely a meaningful proposition.

A tube furnace for the indirect heating of gases or liquids is described in Austrian patent specification No 235 802. The fluid is passed through counterflow tubes, that is to say double tubes which are fitted one into the other, and which project oriented in mutually parallel relationship into a firing chamber. The tubes can have a catalyst filling. This arrangement can provide for a good transfer of heat from the firing chamber to the counterflow tubes. Minimisation of heat losses of the reaction regions in the counterflow tubes to the surrounding area however cannot be achieved therewith.

British patent specification No 1 122 426 sets forth a method of carrying out in particular gaseous phase reactions with simultaneous avoidance of secondary reactions by limiting the temperature of the walls of the reaction region. The educt stream is guided to a reaction region, while the product stream is discharged over the length of the reaction region in counterflow relationship with the educt stream and in thermal contact therewith. That provides for a continuous dissipation of heat so that the walls of the reaction region do not assume such a temperature which causes unwanted catalytic activity. Good transfer of heat from the

product stream to the educt stream is not achieved by virtue of the short length and the arrangement of the counter-flow region. On the contrary, high levels of heat loss are involved by virtue of the heated educt stream and the product stream being guided in wide regions in contact with the surrounding area.

Therefore the object of the present invention is to provide a method of carrying out chemical reactions in a microreactor, and a corresponding microreactor, of the kind set forth in the opening part of this specification, in which heat losses from the reaction region to the surrounding area are greatly reduced and wherein moreover a compact arrangement is possible.

That object is attained by a method as set forth in claim 1 and by a microreactor as set forth in claim 7, while the appendant claims concern advantageous considerations of the invention.

In accordance with the method of the invention the educt stream and the product stream or the educt streams and the product streams are guided spirally or radially in at least one plane respectively to or from a reaction region arranged in a central region of the microreactor, for the purposes of minimising thermal losses from the reaction region to the surrounding area.

The fact that the product stream is guided in counterflow relationship and in thermal contact with the educt stream provides for recovery of the reaction heat contained in the product stream. The invention however also provides for minimisation of the heat losses of the reaction region to the surrounding area, which are very high precisely in the case of microreactors by way of the housing because of the high surface-to-volume ratio. For that purpose the reaction region is disposed in a central region of the microreactor and surrounded in at least one plane spirally or radially by the one or more educt and product streams. Reaction heat which is liberated is therefore fed to the reaction region again by way of the incoming educts or is given off to the product stream which, by virtue of the counterflow relationship, feeds the reaction heat to

the educt stream again. The losses from the reaction region to the surrounding area due to heat conduction or heat radiation, in particular by way of the housing, are thus kept at a minimum. The method according to the invention can also advantageously be used for liquid-phase reactions.

Minimisation of the heat losses means that it is possible to achieve in the reaction region a high temperature which cannot readily be achieved for known microreactors, so that this method crucially expands the area of use of microreactors. Therefore, with this method, the advantages which are linked to the use of microreactors such as an increase in selectivity and yield can be expanded to a new range of chemical reactions. The invention is therefore particularly suitable for carrying out gaseous phase reactions at elevated temperatures, preferably at temperatures of over 500°C.

This method makes it possible to eliminate expensive cooling of the products or heating of the educts, whereby a noticeable energy saving is also achieved. Pre-heating of the educt stream can nonetheless be provided to compensate for remaining heat losses.

If a plurality of reaction regions are involved, they are in adjacent relationship and surrounded by the feed and discharge means. That adjacent arrangement of the reaction regions permits homogenisation of the reaction temperatures and, when using catalysts, homogenisation of the catalyst temperatures. In that way the reactions can be carried out in the microreactor under uniform conditions. Furthermore the arrangement of feed means, discharge means and reaction region permits a compact structure with a simple design configuration so that inexpensive implementation in large numbers is made possible.

In accordance with a first embodiment of the method an educt and a product stream are fed spirally in one plane respectively to and from a reaction region arranged at the common axis of the spiral. To avoid heat losses in the direction above and below the plane of the spiral, the educt and product streams advantageously have a high aspect ratio, that is to say the ratio of height to width, and/or a plurality of educt and product

streams are guided spirally in mutually superposed planes. It is also possible for two or more educt and/or product streams to be guided parallel in one plane. Likewise it is also possible to provide two or more reaction regions which are possibly arranged in mutual succession.

In accordance with a second embodiment a plurality of educt and product streams are respectively guided radially to or from the central region respectively in alternate sectors of a space which is of an annular configuration in cross-section and which surrounds a cylindrical central region, wherein the central region and/or the region of the sectors serves or serve as a reaction region. In this arrangement therefore the reaction region is radially surrounded in at least one plane by the educt and product streams which are guided in the sectors in counterflow relationship with each other.

In accordance with a third embodiment a plurality of educt and product streams are guided radially respectively to and from a plurality of reaction regions which are fluidically separated from each other and which are in mutually adjacent relationship in a central region of the microreactor. At least one respective educt and product stream are guided in a counterflow reactor unit in thermal contact with each other and in counterflow relationship with each other, wherein the counterflow reactor unit respectively has at least one reaction region. The counterflow reactor units are therefore adjacent in at least one plane and oriented towards a central region so that the reaction regions are also in mutually adjacent relationship. The educt and product streams extend radially in the direction of the central region. The heat which is given off by a reaction region is therefore given off in that plane to the educt and product streams and to the other reaction regions so that homogenisation of the temperatures of the reaction regions and therewith uniform reaction implementation in the counterflow reactor units is achieved.

It may be advantageous to pass a heat exchange medium around the counterflow reactor units. The intermediate spaces between the counterflow reactor units can be used for that purpose.

To divide up an educt stream and to bring individual product streams together, the educt streams and/or the product stream are fed or discharged respectively by way of an outer space or chamber of an annular configuration. That also provides for a further heat exchange effect between the educt and product streams.

In order to ensure good heat transfer, the educt and/or product streams are advantageously guided in passages of a width in at least one region of < 2 mm. Even very small passage widths of 500 μm and smaller can be produced using micro-engineering manufacturing processes, whereby very good heat transfer between the product and educt streams is achieved even with short travel lengths and/or high flow speeds. A further advantage of small passage widths is the presence of laminar flow conditions which, in contrast to turbulent conditions which are otherwise conventional in reactors, permit accurate adjustment of the residence times and thus an increase in both selectivity and also yield.

The microreactor according to the invention is distinguished in that the one or more feed means and the one or more discharge means are arranged spirally or radially in at least one plane around the reaction region which is in a central region of the microreactor. The fact that the reaction region is surrounded in at least one plane by the one or more feed means and discharge means respectively provides for minimising heat losses from the reaction region to the surrounding area.

In order to minimise heat losses perpendicularly to the plane in which the reaction region is surrounded by the feed and discharge means, the feed and discharge means advantageously have a high aspect ratio, that is to say ratio of height to width, and/or a plurality of such microreactors are arranged in mutually superposed relationship in such a way that the reaction regions are disposed in mutually adjacent relationship.

In accordance with a first embodiment of the microreactor the feed and discharge means extend spirally around a common spiral axis, wherein the reaction region is arranged in a region at the common spiral

axis. Two or more feed means can also be provided for introducing two or more educts. Two or more reaction regions which are possibly arranged in mutual succession may equally advantageously also be provided.

In accordance with a second embodiment the microreactor has a cylindrical central region which is surrounded by a space or chamber which is divided into sectors and which is of an annular configuration in cross-section. Alternate sectors of the space or chamber of an annular configuration serve as feed means and discharge means respectively. The central region and/or regions of the sectors serves or serve as a reaction region. The sectors which serve as the feed means are respectively connected by way of the central region to the sectors serving as the discharge means. For that purpose the cylindrical central region can simply be in the form of a free space into which the sectors open, or the central region is subdivided in such a way that only one respective sector is connected to its adjacent sector.

In accordance with a third embodiment a plurality of feed and discharge means are arranged radially in at least one plane around a plurality of reaction regions which are fluidically separated from each other and which are disposed in mutually adjacent relationship in a central region of the microreactor. At least one respective feed means and discharge means and at least one respective reaction region are contained in a counterflow reactor unit.

The counterflow reactor units are therefore disposed in mutually adjacent relationship in at least one plane and are oriented towards the central region in such a way that the reaction regions are in mutually adjacent relationship and the feed and discharge means are arranged radially around the reaction regions.

In accordance with a configuration of this embodiment the counterflow reactor units each have a counterflow tube which includes an outer tube closed at one end and an inner tube spaced coaxially in relation to the outer tube. The inner tube serves as a feed or discharge means and the region between the outer tube and the inner tube serves conversely

as a discharge means or feed means respectively. The transitional region between the feed means and the discharge means in the region of the closed end of the outer tube serves as the reaction region. Those counterflow tubes are arranged radially around a central region in such a way that the reaction regions are disposed in mutually adjacent relationship and are oriented towards the central region.

To minimise heat losses perpendicularly to the plane of the counterflow reactor units, there are advantageously provided two or more mutually superposed planes with counterflow reactor units, which are respectively arranged in a plane radially around a common cylindrical central region. Such an arrangement is technically easy to carry into effect.

It is also possible for the counterflow reactor units to be arranged radially in the space around a spherical central region. Such an arrangement which for example is spherically symmetrical substantially prevents direct heat losses of the reaction regions to the surrounding area.

In accordance with a further configuration the counterflow reactor units respectively have two mutually spaced plate-shaped elements which form a passage serving as a feed means or discharge means, and a plate-shaped element which surrounds the two plate-shaped elements in a U-shaped configuration and which, with the first two plate-shaped elements, forms a respective passage serving as a discharge or feed means respectively. The transitional region between the feed and discharge means in the region of the bend of the U-shaped element serves as the reaction region.

Those counterflow reactor units, by virtue of the plate-shaped elements being of almost any desired extent, permit very high aspect ratios and thus very good heat exchange conditions, while involving very low levels of heat loss to the surrounding area.

Due to the use of plate-shaped elements which are in part bent or curved, the microreactor is of a simple structure and can thus be produced

inexpensively. In particular the U-shaped plate-shaped elements of the individual counterflow reactor units, as they are mutually adjoining, can be produced from a single piece.

In accordance with one of the alternative configurations the counterflow reactor units can be mutually adjoining in order to achieve homogenisation of the temperature of the reaction regions by heat conduction. If there are intermediate spaces between the counterflow reactor units, they can be used for passing a heat exchange medium therethrough.

The sectors in accordance with the second embodiment or the counterflow reactor units in accordance with the third embodiment are advantageously surrounded by an outer space or chamber which is of an annular configuration in cross-section and which serves as a discharge collector or manifold or a feed distributor or manifold and which is connected in the radial direction to the discharge means and to the feed means respectively. That permits improved heat exchange with at the same time a compact structure.

The above-mentioned space of an annular configuration, which serves as a discharge collector or a feed distributor can be surrounded by a second outer space or chamber which is of an annular configuration in cross-section and which conversely serves as a feed distributor or discharge collector respectively and which for that purpose is connected in the radial direction to the feed means or the discharge means respectively.

A main discharge means or a main feed means can be provided above and/or below the space in the form of the circular ring, which is divided into sectors, and/or the first and/or second outer space in the form of the circular ring. For that purpose the main feed means or the main discharge means is connected in the axial direction to the sectors serving as the discharge means or the feed means respectively, or the discharge collector or the feed distributor respectively. Besides the extremely compact design configuration, this arrangement minimises heat

losses perpendicularly to the plane in which the feed and discharge means are arranged.

The feed and/or discharge means are advantageously in the form of passages of a width in at least one region of < 2 mm, for good heat transfer. Passage widths of less than or equal to 500 μm are preferred, whereby very good heat transfer between product and educt stream is achieved even with short travel lengths and/or high flow speeds. In a particularly preferred feature the passage widths are so selected that laminar flow conditions prevail, presuming corresponding volume flows.

Particularly for implementing heterogeneously catalysed gaseous phase reactions, the reaction region has a catalytically active material. This can be applied to a carrier material or the walls surrounding the reaction region can have a catalytically active material, for example they may consist thereof or they may have it as a coating.

The microreactors can be produced from individual, suitably bent plate-shaped parts such as sheets or foils, or from a substrate, into which suitable structures such as passages and cavities are introduced, which are possibly closed off by a cover plate.

Suitable production processes are in particular precision-engineering and micro-engineering processes such as milling, spark erosion, ablation by means of laser radiation, lithographic processes using etching procedures, in particular wet etching of high-quality steels, or LIGA-processes. Inexpensive mass production can also be effected with casting or molding processes such as injection molding or die casting or stamping. In this respect, besides polymers, it is also possible to use pre-ceramic compounds or organically bonded metallic and/or ceramic powders, in which case the method possibly requires a bonding agent-removal step and a heat treatment.

Suitable materials for such microreactors are selected in particular having regard to chemical inertness in respect of the educts and the products, temperature stability and workability. Depending on the respective use involved therefore consideration is given to in particular

metals, high-quality steels, alloys, semi-metals (metalloids) such as silicon, glass, ceramic or polymers such as thermoplastic materials.

Further functional units such as mixers, heat exchangers or sensors, for example quantitative flow rate meters, pH-probes, pressure or temperature measuring devices may be integrated into such microreactors.

Embodiments of the microreactor according to the invention are described in greater detail hereinafter with reference to the accompanying diagrammatic drawings in which:

Figure 1a is a view in cross-section from above showing a microreactor with a spiral arrangement of the feed and discharge means,

Figure 1b is a side view in cross-section of the microreactor shown in Figure 1a,

Figure 2 is a view in cross-section from above showing a microreactor with a spiral arrangement of two feed means and a discharge means,

Figure 3a is a view in cross-section from above showing a microreactor with a space of an annular configuration, which is divided into sectors serving as feed means and discharge means respectively,

Figure 3b is a side view in cross-section of the microreactor shown in Figure 3a,

Figure 4a is a view from above in cross-section showing a microreactor with radially arranged counterflow reactor units in the form of counterflow tubes,

Figure 4b is a side view in cross-section of the microreactor shown in Figure 4a,

Figure 5a is a view from above in cross-section showing a microreactor with radially arranged counterflow reactor units formed from plate-shaped elements,

Figure 5b is a side view in cross-section showing the microreactor of Figure 5a, and

Figure 5c is a perspective view in section from above and partly from the side of the microreactor shown in Figures 5a and 5b.

Referring to Figure 1a, shown therein is a diagrammatic view from above in cross-section of a microreactor 1 with a spiral arrangement of the feed means 2 and the discharge means 3. The feed means 2 and the discharge means 3 are guided in adjacent relationship in one plane in the manner of a double spiral and both are connected to the reaction region 4 which is in the common central region 4'. The arrows indicate the direction of flow of the educt and product stream respectively within the microreactor 1. Except for the outer turn, the feed means 2 is surrounded in the illustrated plane by the discharge means 3. Because the reaction region 4 is surrounded in the illustrated plane by the feed and discharge means 2, 3, heat losses to the surrounding area are greatly minimised in that plane. The adjacent arrangement and fluid guidance in counterflow relationship permit effective heat exchange between the product stream and the educt stream. A catalyst material 5 is disposed in the reaction region 4.

Figure 1b is a side view in cross-section of the microreactor illustrated in Figure 1a. The adjacent arrangement of the feed means 2 with the discharge means 3 which are both connected to the reaction region 4, here with a catalyst material 5, is clearly visible herein. In order to reduce heat losses to the top and bottom plates 6, 7 which close off the feed means and the discharge means at the two end faces, the feed means 2 and the discharge means 3 have a high ratio between height and width. It is also possible for a plurality of such microreactors to be stacked one upon the other whereby there are no heat losses to the adjacent closing elements.

The microreactor shown in Figure 2 has two feed means 10 and 11 and a discharge means 12 which are arranged around the reaction region 13 in the central region 13' and are connected thereto. It is also possible to provide three or more feed means or two or more reaction regions which are possibly arranged in mutual succession.

Such microreactors which are shown in Figures 1a, 1b and 2 can be made up from plate-shaped elements which are wound in a spiral configuration and which are closed at the end faces, and have connections for the feed and the discharge means. It is also possible for the walls of the feed and the discharge means and a bottom plate to be produced in one piece from a substrate, for example by microstructuring of a polymer and subsequent galvanic molding or by stamping of an aluminum plate.

Figure 3a diagrammatically shows a microreactor 20 having a space or chamber 29a which is of an annular configuration in cross-section and which surrounds a cylindrical central region 30 and which is divided into sectors 31a, b. Alternate sectors 31a, b, here in each case seventeen, serve as feed means 22 and as discharge means 23 respectively. The sectors 31a serving as feed means 22 are connected by way of the central region 30 which is formed by a free space to the sectors 31b serving as discharge means 23. In this example, the regions, which are towards the central region 30, of the sectors 31b serving as the discharge means 23 serve as reaction regions 21. The space 29a of an annular configuration has a plurality of sectors, thereby providing for a good heat exchange effect between the product and educt streams which are guided in counterflow relationship and which flow radially to and from the central region respectively. In this example, the sectors 31a serving as feed means 22 are in the form of passages of approximately equal width while the sectors 31b serving as discharge means 23 are of a cross-section which enlarges radially outwardly. The walls 24 which delimit the feed and discharge means 22, 23 and which form the sectors 31a, b are advantageously formed from a material which is a good conductor of heat, for example metal foils or sheets. The space 29a of an annular configuration is surrounded by an outer space 29b which is of an annular configuration and which serves as a feed distributor 27a. For that purpose, it is connected to a laterally arranged main feed means 27b and by way of slots 29c to the sectors 31a serving as feed means 22. The directions of flow of the educt and product streams are indicated as arrows. In this

example, the reaction regions 21 have a catalytically active material 25 which is applied to a carrier 26. It may also be advantageous to provide a catalytically active material in the central region 30. In order to achieve a change in temperature, for example when starting a reaction in the microreactor, in the central region 30 of the microreactor 20, it may be advantageous to arrange a heating or cooling device, for example a heat exchanger, in the central region 30.

The lateral cross-section shown in Figure 3b of the microreactor 20 illustrated in Figure 3a also indicates the configuration of the educt and the product streams by means of arrows. Also shown therein is a reaction region 21 with a catalytic material 25 which is applied to the carrier 26 and which is connected by way of the feed means 22, the feed distributor 27a to the lateral main feed means 27b and by way of the discharge means 23 to the main discharge means 28. The main discharge means 28 are arranged at both ends of the microreactor 20 above and below both the central region 30, the space 29a which is of an annular configuration and which is divided into sectors, and also the outer space 29b which is also of an annular configuration, and are axially connected to the sectors 31b serving as the discharge means 23.

Figure 4a shows a view from above in cross-section of a microreactor 40 which in the illustrated plane has five counterflow reactor units 41. Each microreactor unit 41 includes a feed means 43 which is formed by an inner tube 44 and which is surrounded in the illustrated plane by the discharge means 45 in the form of an outer tube 42, and is connected thereto by way of the reaction region 46 which is disposed at the closed end of the outer tube 42. The individual counterflow reactor units 41 are arranged radially in such a way that the reaction regions 46 which are not in fluidic communication with each other are disposed in adjacent relationship in the central region 46'. In this example the region 46' between the counterflow reactor units 41 is empty, whereby heat transfer to adjacent counterflow reactor units takes place predominantly by heat radiation. The adjacent arrangement of the reaction regions 46

minimises heat transfer to the surrounding area. It is also possible to use the free region 46' for passing a heat exchange medium therethrough, for example in the starting phase of a reaction, for temperature regulation purposes or to dissipate excess heat. The discharge means 45 of the counterflow reactor units 41 are connected to a discharge collector or manifold 49a which is in the form of a space or chamber 49b of an annular configuration. The feed means 43 begin in a common feed distributor 47a which in the form of a second outer space or chamber 47b of an annular configuration surrounds the discharge collector 49a.

Figure 4b is a side view in cross-section showing the microreactor 40 illustrated in Figure 4a. The counterflow reactor units 41 each include an outer tube 42 and a coaxially arranged inner tube 44, wherein both tubes are of a round cross-section. The outer tube 42 is hemispherically closed at one end. The inner tube 44 forms the feed means 43 which is surrounded by the discharge means 45. The reaction region 46 is disposed in the transitional region between the feed means 43 and the discharge means 45, preferably in the region of the closed end of the outer tube 42. The counterflow reactor units 41 are arranged in the interior of the discharge collector 49a of an annular configuration, radially in three planes around the cylindrical central region 46'. The discharge collector 49a connects the individual discharge means 45 to the main discharge means 49c which are arranged at both end faces of the cylindrical microreactor 40. The feed distributor 47 which is in the form of the second outer space 47b of an annular configuration communicates the main feed means 48 with the individual feed means 43. The microreactor units 41 are arranged radially around the central region 46' in such a way that the reaction regions 46 are adjacent. By virtue of greatly reduced heat radiation losses to the surrounding area, both effective concentration and also homogenisation of the reaction heat in the central region 46' of the microreactor 40 is possible. A large part of the reaction heat contained in the product stream can be given off to the educt stream in the region of the feed means 43 and the feed distributor 47.

The microreactor 40 can be made up of three hollow cylinders 80a, b, c which are disposed coaxially one within the other, for example consisting of metal foil or sheet. The outer tubes 42 of the counterflow reactor units 41 in this case represent protuberances of the innermost hollow cylinder 80a. The inner tubes 44 of the counterflow reactor units, which are connected to the central hollow cylinder 80b, are not in contact with the outer tubes 42 surrounding them, so that even major temperature differences do not result in internal stresses in the hollow cylinder arrangement 80a, b, c, by virtue of that free mobility.

The microreactor 50 diagrammatically shown in Figures 5a, 5b and 5c has five counterflow reactor units 51 which surround a central region 60' in such a way that the reaction regions 60 of the individual counterflow reactor units 51 are in mutually adjacent relationship in the central region 60'. Each counterflow reactor unit 51 has a feed means 56 which is surrounded by the discharge means 58 in the plane shown in Figure 5a, wherein the connecting region between the feed and the discharge means forms the reaction region 60. The educt and product streams which are carried by the feed means 56 and the discharge means 58 respectively and the configuration of which is here indicated by arrows go radially to and from the reaction regions 60 in the central region 60'. The counterflow reactor units 51 are surrounded by a space 59b which serves as a discharge collector 59a and which is of an annular configuration and which is connected to the discharge means 58. The discharge collector 59a in turn is surrounded by a feed distributor 57a which is in the form of a second outer space 57b of an annular configuration, and which distributes the educt stream to the individual feed means 56, from a main feed means 61 arranged laterally on the microreactor 50. Disposed above and below the cylindrical microreactor 50 is a main discharge means 62 which discharges the products axially from the discharge collector 59a.

Each feed means 56 of a counterflow reactor unit 51 is formed by two mutually spaced plate-shaped elements 54, 55. They are surrounded by a U-shaped plate-shaped element 53 in such a way that formed in the

two intermediate spaces between the elements 54 and 55 respectively and the U-shaped element 53 is a respective passage which serves as a discharge means 58. The transitional region between the feed means 56 and the two discharge means 58, in particular the region of the bend of the U-shaped element 53, serves as a reaction region 60. It will be clearly apparent in particular from Figure 5c that, in this embodiment, the U-shaped elements 53 of the five counterflow reactor units 51 are formed from a single, suitably shaped plate-shaped element which represents a hollow cylinder 90a with U-shaped protuberances. Each two plate-shaped elements 54, which form a respective wall of a feed means 56, of two adjacent counterflow reactor units 51 are formed from a plate-shaped element. Those plate-shaped elements also form the wall, identified as the central hollow cylinder 90b, between the space 59b which is of an annular configuration and which serves as a discharge collector 59a, and the second outer space 57b which is also of an annular configuration and which serves as a feed distributor 57a. The outer hollow cylinder 90c forms the outer wall of the feed distributor 57a. As can be seen from Figure 5c, the hollow cylinders 90a, b, c are connected to a bottom plate 63 which has openings 64 for discharging the product streams from the discharge collector 59a into the main discharge means 62. In a corresponding manner the connection is constituted by a cover plate (not shown here) in relation to the main discharge means 62 which is arranged above. This compact microreactor 50 comprises a small number of elements which are simple to produce, and it can thus be used as an inexpensive mass-produced article in many areas, for example in fuel cell technology and/or motor vehicle technology.

The microreactors are of an extremely compact structure, in which respect the reaction regions are surrounded in at least one plane by educt and product streams respectively. The counterflow configuration provides that heat which is given off by the reaction regions is fed to same again so that markedly higher reaction temperatures can be achieved in comparison with known microreactors. Therefore these microreactors

afford a further range of uses which hitherto were not considered for microreactors, in particular in terms of chemical reactions at high temperatures.

List of references

1	microreactor
2	feed means
3	discharge means
4	reaction region
4'	central region
5	catalyst material
6	top plate
7	bottom plate
10	feed means
11	feed means
12	discharge means
13	reaction region
13'	central region
20	microreactor
21	reaction region
22	feed means
23	discharge means
24	wall
25	catalyst material
26	catalyst carrier
27a	feed distributor
27b	main feed means
28	main discharge means
29a	space of an annular configuration
29b	outer space of an annular configuration
29c	slot
30	cylindrical central region
31a, b	sector
40	microreactor
41	counterflow reactor unit
42	outer tube

43	feed means
44	inner tube
45	discharge means
46	reaction region
46'	central region
47a	feed distributor
47b	second outer space of an annular configuration
48	main feed means
49a	discharge collector
49b	outer space of an annular configuration
49b	main discharge means
50	microreactor
51	counterflow reactor unit
53	plate-shaped element bent in a U-shape
54	plate-shaped element
55	plate-shaped element
56	feed means
57a	feed distributor
57b	second outer space of an annular configuration
58	discharge means
59a	discharge collector
59b	outer space of an annular configuration
60	reaction region
60'	central region
61	main feed means
62	main discharge means
63	bottom plate
64	opening
80a, b, c	hollow cylinder
90a, b, c	hollow cylinder

CLAIMS

1. A method of carrying out chemical reactions in a microreactor, in which one or more educt streams are guided to at least one reaction region and in which one or more product streams are guided from the reaction region in counterflow relationship with the educt stream or streams and in thermal contact therewith, characterised in that to minimise heat losses from the reaction region to the surrounding area the educt stream or streams and the product stream or streams are guided spirally or radially in at least one plane respectively to or from the reaction region arranged in a central region of the microreactor.

2. A method as set forth in claim 1 characterised in that a plurality of educt and product streams are respectively guided radially to and from the central region in alternate sectors of a space which is of an annular configuration in cross-section and which surrounds a cylindrical central region, wherein the central region and/or regions of the sectors serves or serve as the reaction region.

3. A method as set forth in claim 1 characterised in that a plurality of educt and product streams are guided radially respectively to and from a plurality of reaction regions which are fluidically separated from each other and which are disposed in mutually adjacent relationship and in a central region of the microreactor, wherein in each case at least one educt and product stream are guided in a counterflow reactor unit which has at least one respective reaction region.

4. A method as set forth in claim 3 characterised in that a heat exchange medium is passed around the counterflow reactor units.

5. A method as set forth in one of claims 2 through 4 characterised in that the educt streams and/or the product streams are fed and

discharged respectively by way of an outer space of an annular configuration.

6. A method as set forth in one of claims 1 through 5 characterised in that the educt stream or streams and/or the product stream or streams are guided in passages of a width in at least one region of < 2 mm.

7. A microreactor (1; 20; 40; 50) for carrying out chemical reactions comprising one or more feed means (2; 10, 11; 22; 43; 56) for educts and one or more discharge means (3; 12; 23; 45; 58) for products and at least one reaction region (4; 13; 21; 46; 60) connected to the feed and discharge means (2, 3; 10, 11, 12; 22, 23; 43, 45; 56, 58), wherein the discharge means are arranged in counterflow relationship with the one or more feed means and are in thermal contact therewith, characterised in that to minimise heat losses from the reaction region (4; 13; 21; 46; 60) to the surrounding area the feed means (2) and the discharge means (3; 12) or the plurality of feed means (10, 11; 22; 43; 56) and the plurality of discharge means (23; 45; 58) are arranged spirally or radially in at least one plane around the reaction region (4; 13; 21; 46; 60) which is in a central region (4'; 13'; 30; 46'; 60') of the microreactor (1; 20; 40; 50).

8. A microreactor (20) as set forth in claim 7 characterised by a cylindrical central region (30) and a space (29a) which is of an annular configuration in cross-section and which surrounds it and which is divided into sectors (31a, b),

wherein alternate sectors (31a, b) of the space of an annular configuration serve as feed and discharge means (22, 23) respectively,

wherein the sectors (31a) serving as a feed means (22) are connected by way of the central region to the respective sectors (31b) serving as a discharge means (23), and

wherein the central region (30) and/or regions of the sectors (31a, b) serves or serve as a reaction region (21).

9. A microreactor (40; 50) as set forth in claim 7 characterised in that a plurality of feed and discharge means (43, 45; 56, 58) are arranged radially in at least one plane around a plurality of reaction regions (46; 60) which are fluidically separated from each other and which are disposed in mutually adjacent relationship in a central region (46'; 60') of the microreactor (40; 50), wherein at least one respective feed means (43; 56) and discharge means (45; 58) and at least one reaction region (46; 60) are contained in a counterflow reactor unit (41; 51).

10. A microreactor (40) as set forth in claim 9 characterised in that the counterflow reactor units (41) each have a counterflow tube including an outer tube (42) closed at one end and an inner tube (44) spaced coaxially in relation to the outer tube, wherein the inner tube (44) serves as a feed or discharge means (43) and the region between the outer and the inner tube serves as a discharge or feed means (45) respectively, and wherein the transitional region between the feed and the discharge means in the region of the closed end of the outer tube serves as a reaction region (46).

11. A microreactor as set forth in claim 9 or claim 10 characterised in that the counterflow reactor units are arranged in two or more planes radially around a cylindrical central region.

12. A microreactor as set forth in claim 9 or claim 10 characterised in that the counterflow reactor units are arranged in the space radially around the central spherical region.

13. A microreactor (50) as set forth in claim 9 characterised in that the counterflow reactor units (51) each have two mutually spaced plate-shaped elements (54, 55) forming a passage serving as a feed or discharge means (56), and a plate-shaped element (53) which surrounds

the two plate-shaped elements in a U-shape and which forms therewith a respective passage serving as a discharge or feed means (58), wherein the transitional region between the feed and the discharge means in the region of the bend of the U-shaped element serves as a reaction region (60).

14. A microreactor as set forth in one of claims 9 through 13 characterised in that the central region having the counterflow reactor units is adapted for a heat exchange medium to pass therethrough.

15. A microreactor (20; 40; 50) as set forth in one of claims 8 through 14 characterised in that the sectors (31a, b) or the counterflow reactor units (41; 51) are surrounded by an outer space (29b; 49b; 59b) which is of an annular configuration in cross-section and which serves as a discharge collector (49a, 59a) or feed distributor (27a) and which is connected in the radial direction to the discharge means (45; 58) and the feed means (22) respectively.

16. A microreactor (40; 50) as set forth in claim 15 characterised in that the outer space (49b; 59b) which is of an annular configuration and which serves as a discharge collector (49a; 59a) or feed distributor is surrounded by a second outer space (47b; 57b) which is of an annular configuration in cross-section and which serves as a feed distributor (47a; 57a) or discharge collector respectively and which is connected in the radial direction to the feed means (43; 56) and the discharge means respectively.

17. A microreactor (20; 40; 50) as set forth in claims 8 through 16 characterised by a main discharge means (28; 49c; 62) or main feed means which is arranged above and/or below the space (29a) which is of an annular configuration and which is divided into sectors and/or the first and/or second outer space (29b; 49b, 47b; 59b, 57b) which is of an

annular configuration and which for that purpose is connected in the axial direction to the sectors (31b) serving as the discharge means (23) or feed means respectively or the discharge collector (49a; 59a) or the feed distributor respectively.

18. A microreactor as set forth in one of claims 7 through 17 characterised in that the feed and/or discharge means are in the form of passages in at least one region of a width of < 2 mm.

19. A microreactor as set forth in one of claims 7 through 18 characterised by catalytically active material which is possibly applied to a carrier, in the reaction region.

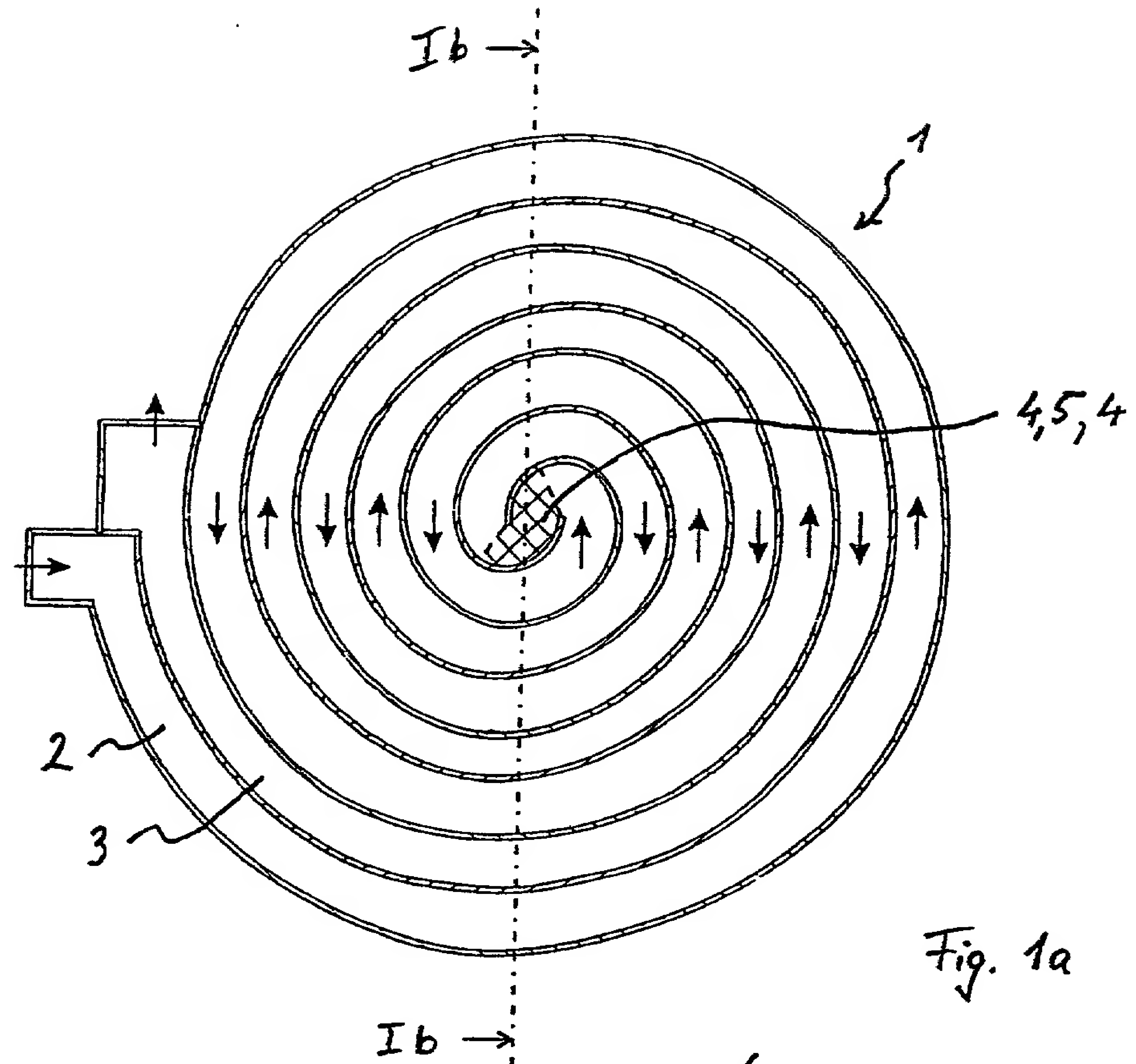

Fig. 1a

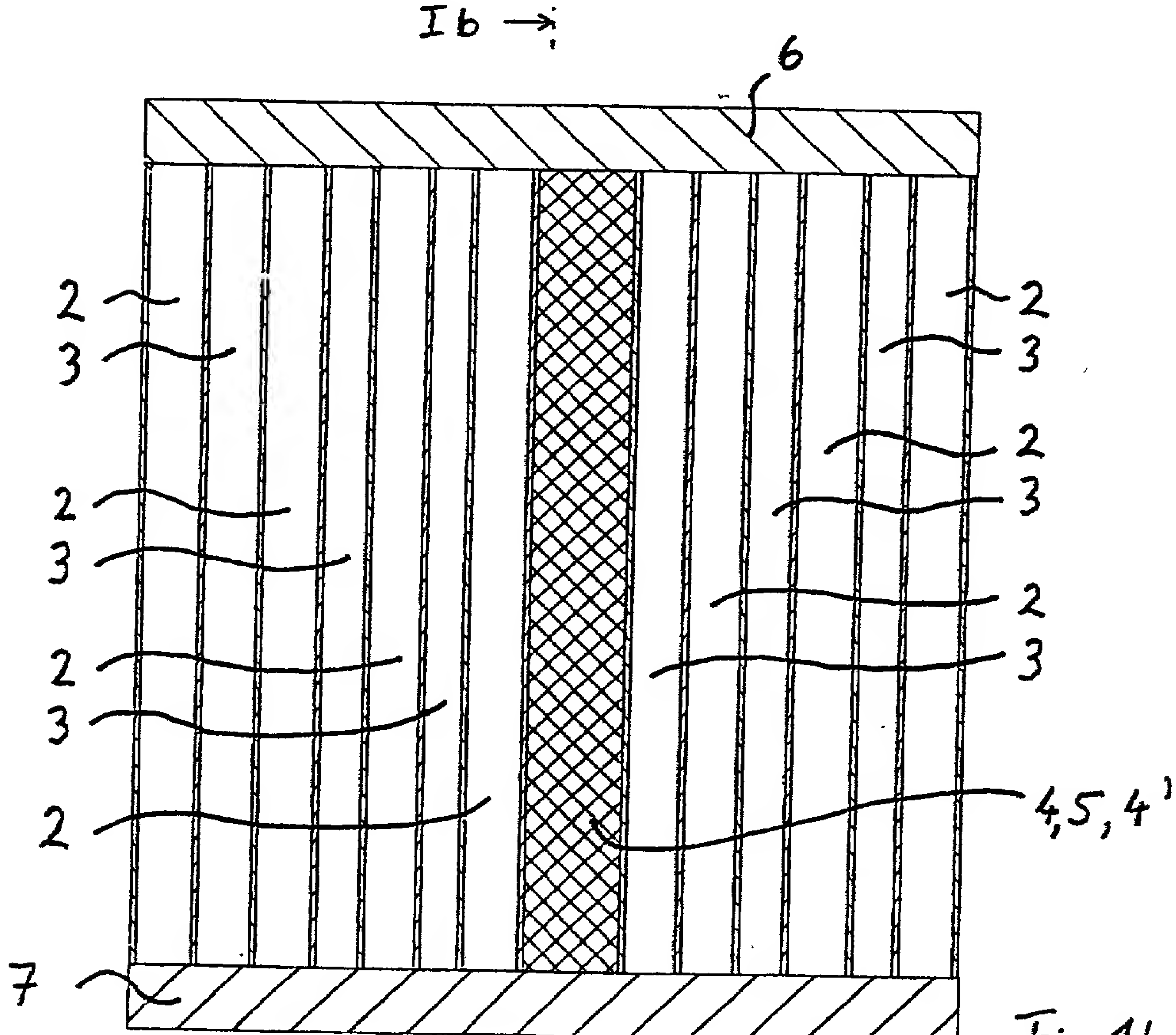

Fig. 1b

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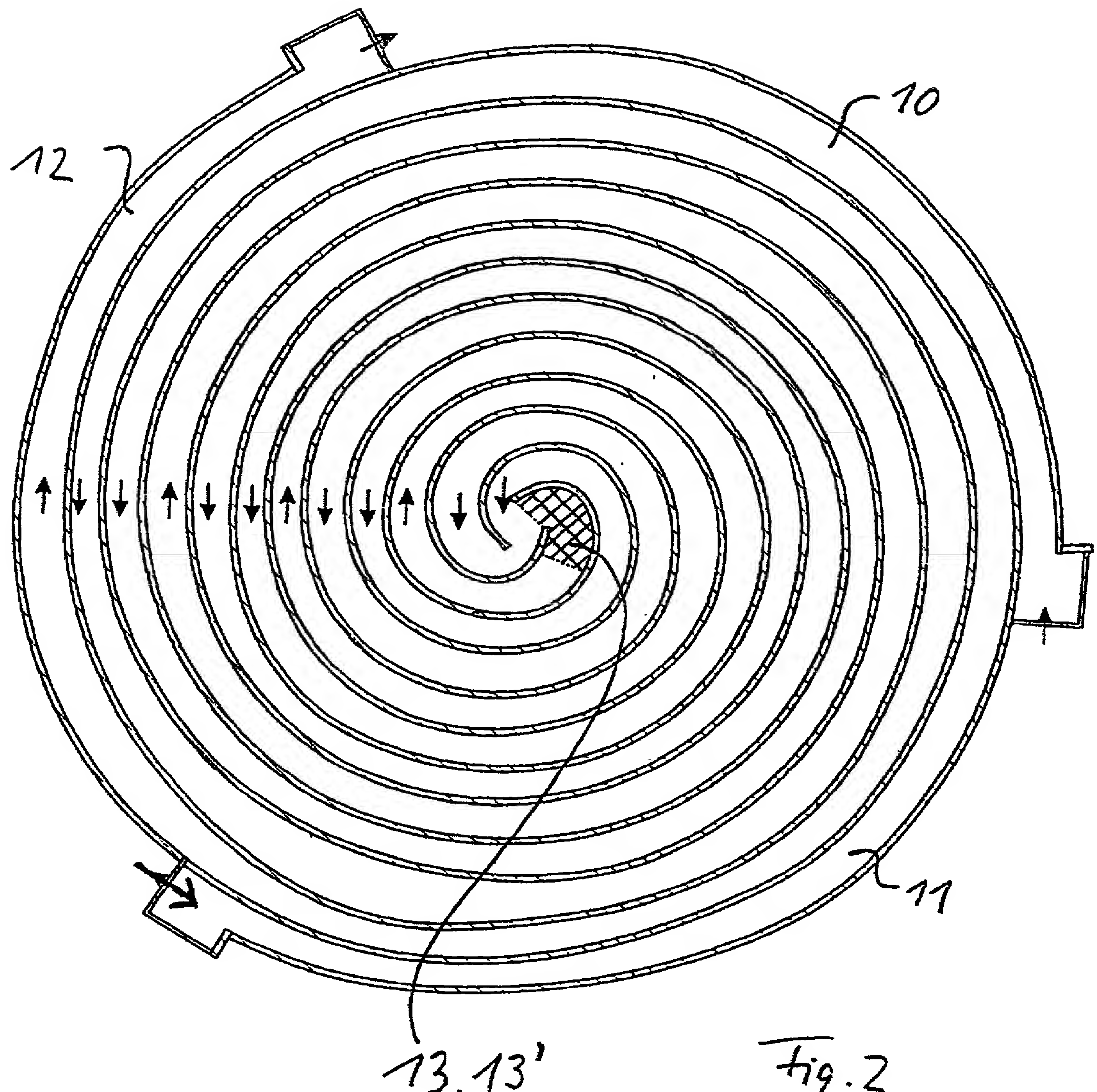

Fig. 2

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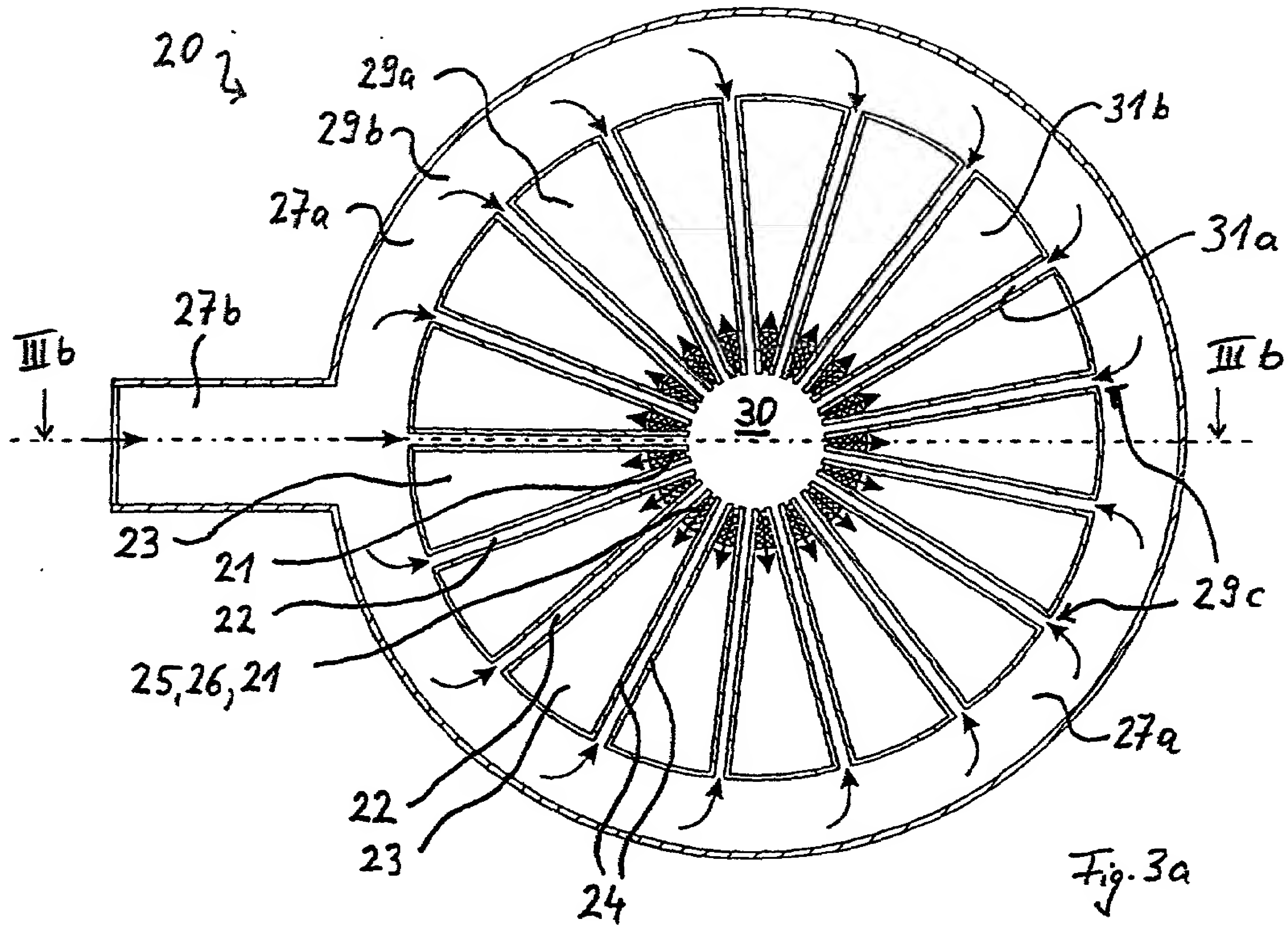

Fig. 3a

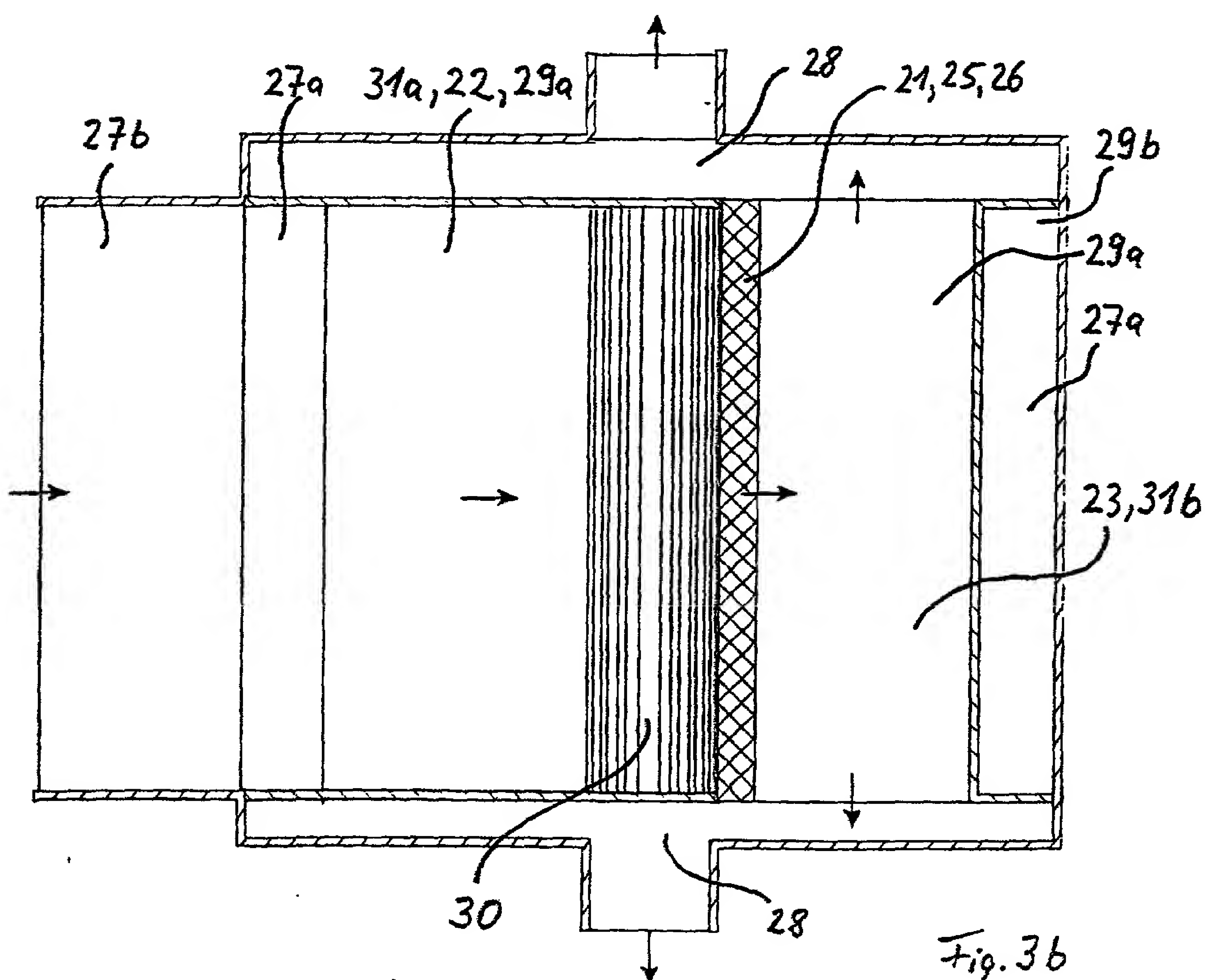

Fig. 3b

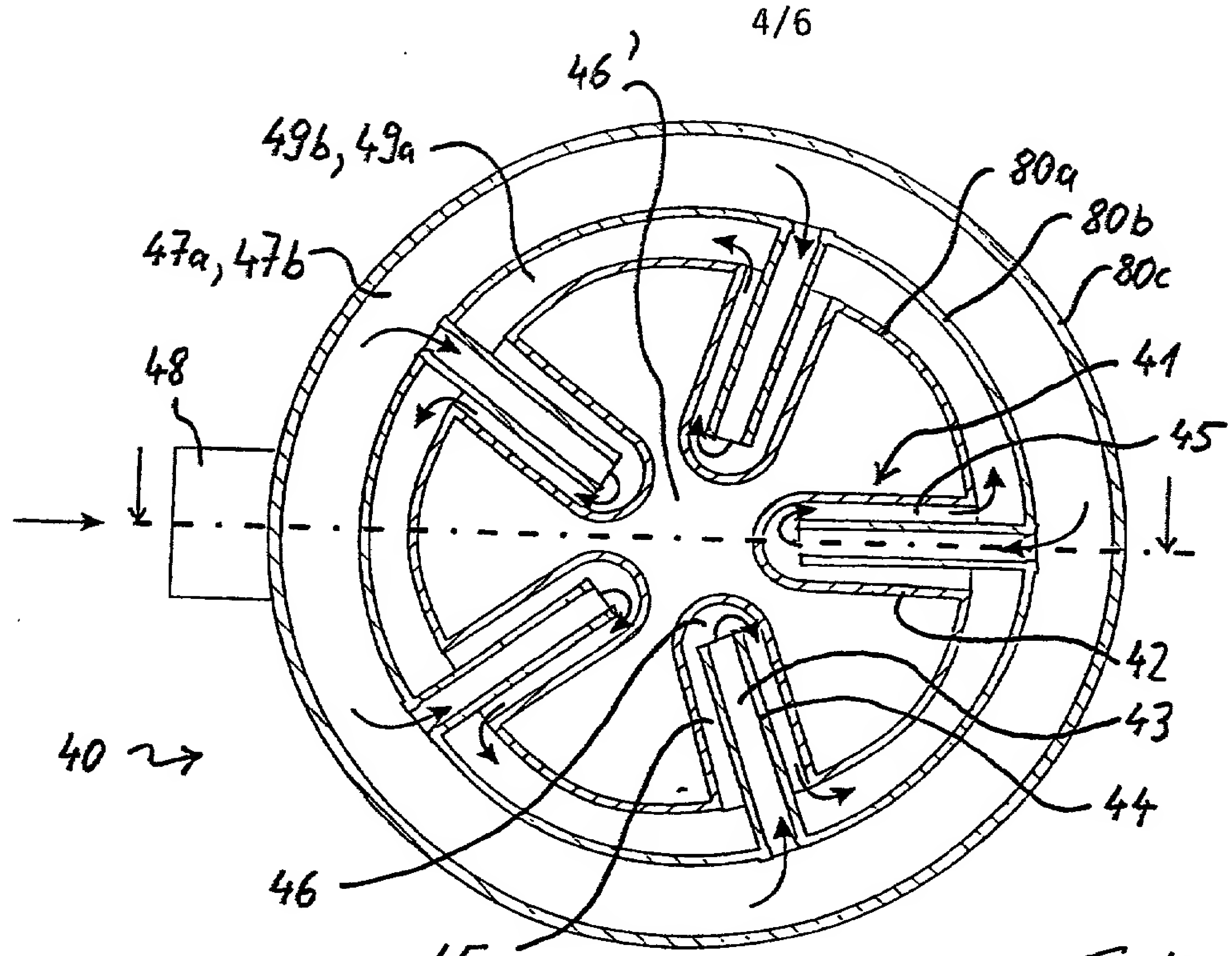
46'
49b, 49a
80a
80b
47a, 47b
80c
48
41
45
42
43
40
44
46
45

Fig. 4a

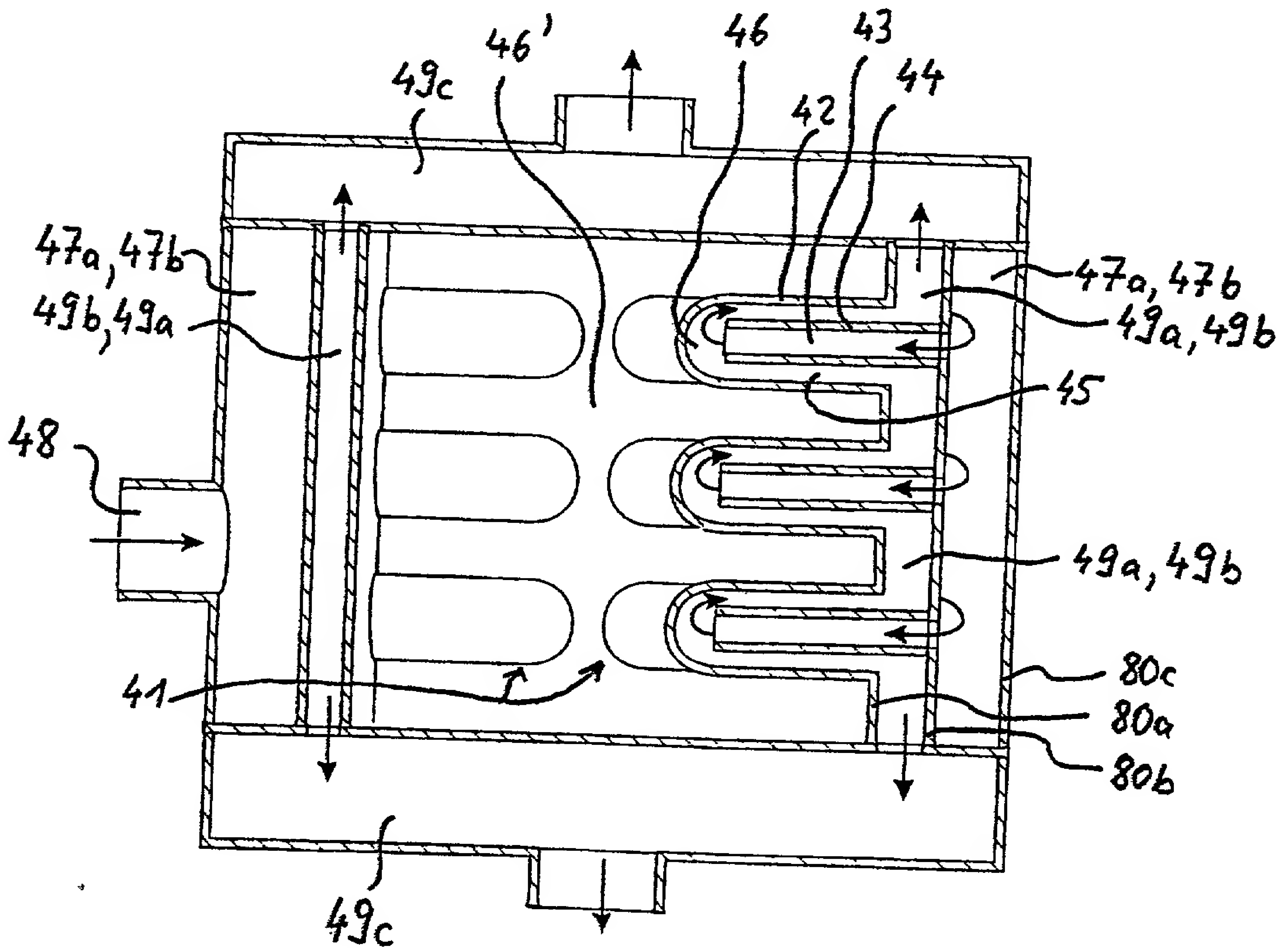
46'
46
43
44
49c
42
47a, 47b
47a, 47b
49b, 49a
49a, 49b
45
48
49a, 49b
80c
41
80a
80b
49c

Fig. 4b

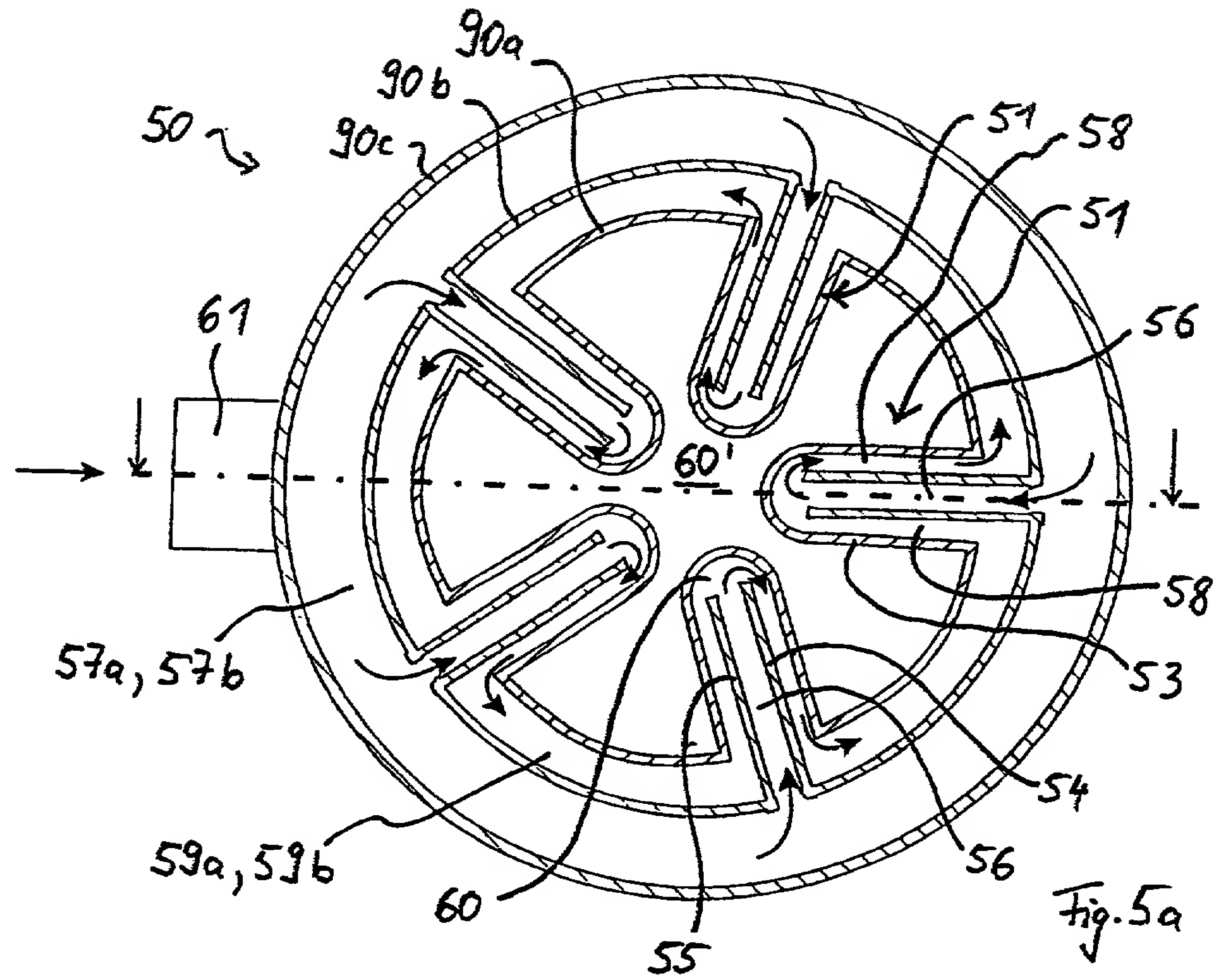
90a
90b
50
90c
51
58
51
56
61
60'
58
53
57a, 57b
54
56
59a, 59b
60
55

Fig. 5a

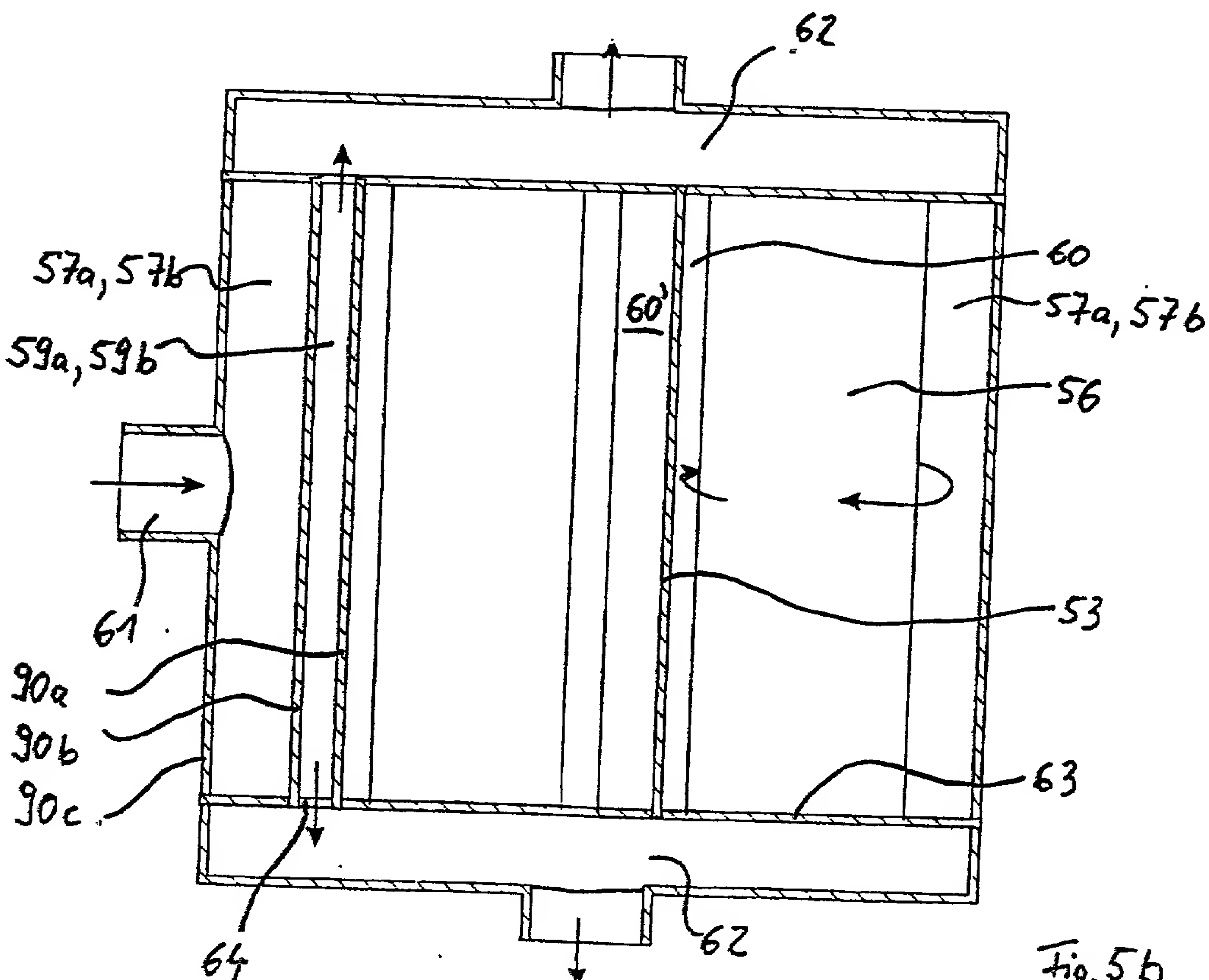
62
60
57a, 57b
60'
57a, 57b
59a, 59b
56
53
61
90a
90b
63
90c
64
62

Fig. 5b

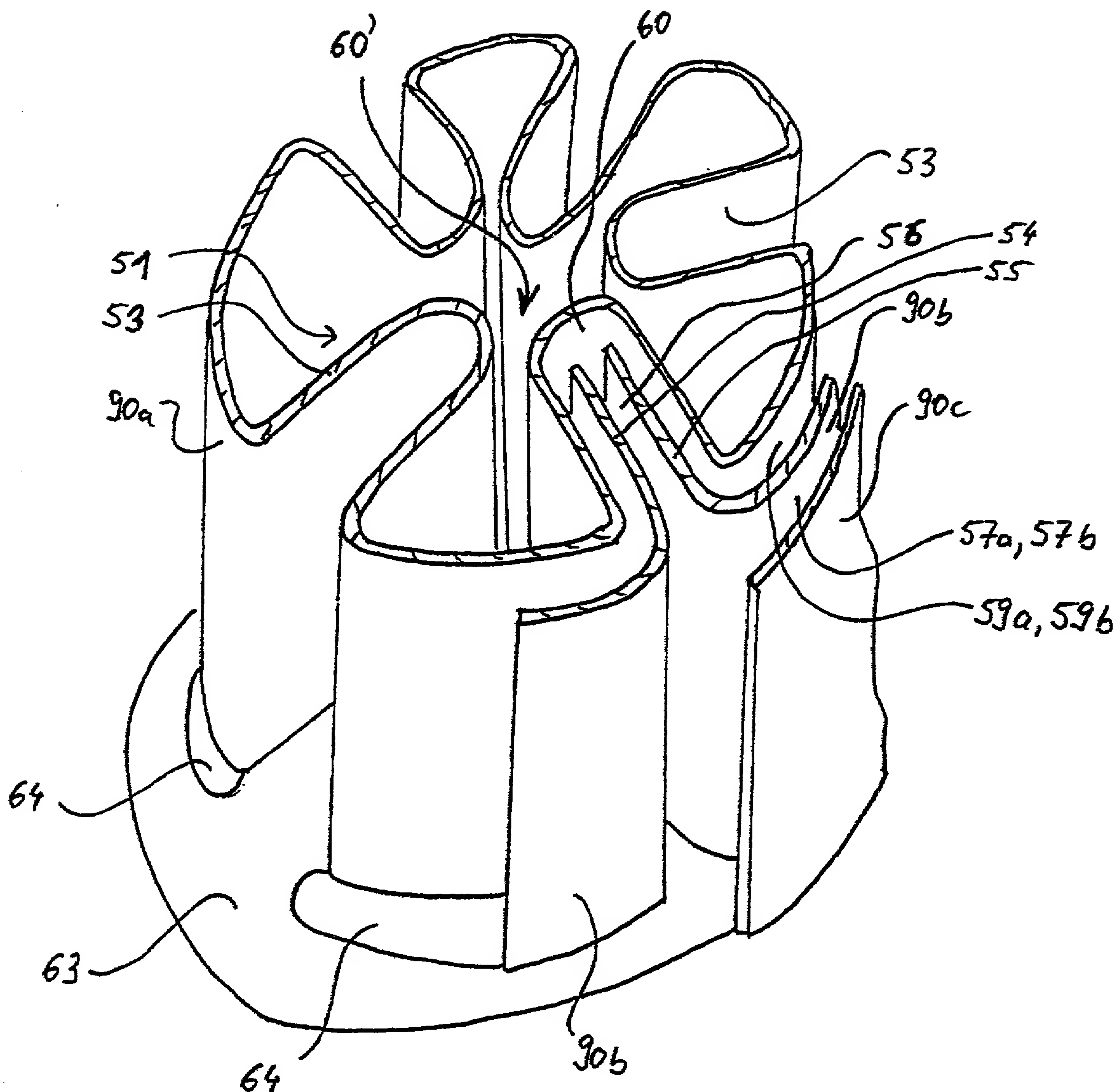

Fig. 5c

EH 75-98

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DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION (37 CFR 1.63)

☐ Declaration Submitted with Initial Filing OR ☐ Declaration Submitted after Initial Filing (surcharge (37 CFR 1.16 (e)) required)

Attorney Docket Number	WEB 0039 PA
First Named Inventor	Wolfgang Ehrfeld
COMPLETE IF KNOWN	
Application Number	/
Filing Date	
Group Art Unit	
Examiner Name	

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

METHOD OF CARRYING OUT CHEMICAL REACTIONS IN A MICROREACTOR AND SUCH A MICROREACTOR

(Title of the Invention)

the specification of which

☐ is attached hereto

OR

XX was filed on (MM/DD/YYYY) 03/03/1999 as ~~United States Application Number or~~ PCT International Application Number PCT/EP99/01361 and was amended on (MM/DD/YYYY) ____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. 119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT international application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? YES	NO
19809139.7	Germany	03/04/1998	☐	☐	☒
			☐	☐	☐
			☐	☐	☐
			☐	☐	☐

☐ Additional foreign application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto:

I hereby claim the benefit under 35 U.S.C. 119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)

☐ Additional provisional application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto.

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PTO/SB/01 (12-97)
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Patent and Trademark Office, U.S. DEPARTMENT OF COMMERCE

DECLARATION — Utility or Design Patent Application

I hereby claim the benefit under 35 U.S.C. 120 of any United States application(s), or 365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application or PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)

☐ Additional U.S. or PCT international application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto.

As a named inventor, I hereby appoint the following registered practitioner(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith: ☐ Customer Number [ ] → Place Customer Number Bar Code Label here

OR

☒ Registered practitioner(s) name/registration number listed below

Name	Registration Number	Name	Registration Number
Richard A. Killworth	26,397	Susan M. Luna	38,769
James F. Gottman	27,262	Patricia L. Prior	33,758
Timothy W. Hagan	29,001	William A. Jividen	42,695
James E. Beyer	39,564	Gregory J. Adams	44,494

☒ Additional registered practitioner(s) named on supplemental Registered Practitioner Information sheet PTO/SB/02C attached hereto.

Direct all correspondence to: ☐ Customer Number or Bar Code Label [ ] OR ☒ Correspondence address below

Name	Richard A. Killworth
Address	Killworth Gottman Hagan & Schaeff, L.L.P.
Address	One Dayton Centre, Sutie 500, One South Main Street
City	Dayton — State OH — ZIP 45402-2023
Country	USA — Telephone 937-223-2050 — Fax 937-223-0724

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor: ☐ A petition has been filed for this unsigned inventor

Given Name (first and middle [if any])	Family Name or Surname
Wolfgang	Ehrfeld

Inventor's Signature	X Wolfgang Ehrfeld			Date	10/15/2000
Residence: City	Mainz	State		Country	Germany DEX
Citizenship	DE				
Post Office Address	Kahlweg 22				
Post Office Address					
City	Mainz	State		Zip	D-55124
Country	DE				

☒ Additional inventors are being named on the supplemental Additional Inventor(s) sheet(s) PTO/SB/02A attached hereto

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DECLARATION	ADDITIONAL INVENTOR(S) Supplemental Sheet Page 3 of 4

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Name of Additional Joint Inventor, if any:	☐ A petition has been filed for this unsigned inventor						
Given Name (first and middle [if any])				Family Name or Surname			
Holger				Löwe			
Inventor's Signature	X Holger Löwe			DEX		Date	10/16/2000
Residence: City	Oppenheim	State		Country	Germany	Citizenship	DE
Post Office Address	Anna-Seghers-Strasse 3						
Post Office Address							
City	Oppenheim	State		ZIP	D-55276	Country	DE

3W

Name of Additional Joint Inventor, if any:	☐ A petition has been filed for this unsigned inventor						
Given Name (first and middle [if any])				Family Name or Surname			
Volker				Hessel			
Inventor's Signature	X Volker Hessel			DEX		Date	09/21/2000
Residence: City	Hünstetten-Wallbach	State		Country	Germany	Citizenship	DE
Post Office Address	Über dem Berg 4						
Post Office Address							
City	Hünstetten-Wallbach	State		ZIP	D-65510	Country	DE

Name of Additional Joint Inventor, if any:	☐ A petition has been filed for this unsigned inventor						
Given Name (first and middle [if any])				Family Name or Surname			
Inventor's Signature						Date	
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		ZIP		Country	

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DECLARATION

REGISTERED PRACTITIONER INFORMATION (Supplemental Sheet)

Name	Registration Number	Name	Registration Number
Thomas E. Lees	P-46,867	//	
John D. Reed	~~P-46,506~~		
Brian L. Smiler	46,458		

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